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CALCULATED RESEARCH ON THE ENERGETIC CHARACTERISTIC OF
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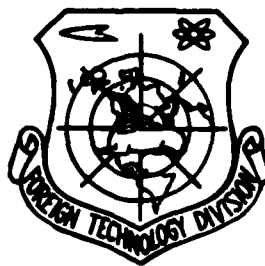
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CALCULATED RESEARCH ON THE ENERGETIC CHARACTERISTIC OF TN IN COMPOSITE
SOLID PROPELLANT

by

Zhang Wei and Tian Deyu



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CALCULATED RESEARCH ON THE ENERGETIC CHARACTERISTIC
OF TN IN COMPOSITE SOLID PROPELLANT

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Zhang Wei & Tian Deyu

ABSTRACT

This paper discussed briefly the properties of a new organic oxidizer--2,3,5,6--Tetranitrato-1,4-Dinitropiperazine (TN). On the basis of the method of White's Minimization of Gibbs Free Energy, we also studied the energetic characteristics of TN in Composite Solid Propellant by means of electronic computer. The calculated results show that the theoretic specific impulse of the system of TN, Al and HTPB reaches 276 seconds, about ten seconds higher than the system of AP, Al and HTPB. This shows that TN does act as a high energetic oxidizer. The paper also did an initial research on the energetic characteristics of composite solid smokeless propellant consisting of TN and HTPB.

1. FOREWORD

Propellants are the sources of power for airborne vehicles. From Oles rocket range formula and terminal velocity of the main-power section equation:

$$v_k = I_s \cdot g \cdot \ln(1 + W_p/W_k) \\ X_s = 2S_s + K \cdot v_k$$

where v_k is the maximum value of the terminal velocity of the main-power section
 I_s is the specific impulse of the propellant
 W_p, W_k are the weight of the propellant and the structural weight of the engine respectively
 S_s is the range for the main-power section
 g is the gravitational acceleration
 K is the constant

From the equations it can be seen that the rocket range is proportional to the specific thrust. On the other hand, when the range and other parameters are fixed, then the increase of specific impulse will reduce the quantity of chemicals. Therefore, the total weight of the engine is reduced. It is because of this that ways and approaches to enhance the energy of propellants represent most important meanings to the development of aero-industries. One of

the major methods to enhance the energy contained in the propellants is to incorporate more energetic materials in the propellants, such as energy-containing oxidizing agents, adhesives, and etc. This article describes the theoretical research on the energetic characteristics of a new energy-containing oxidizing agent TN in the solid propellant.

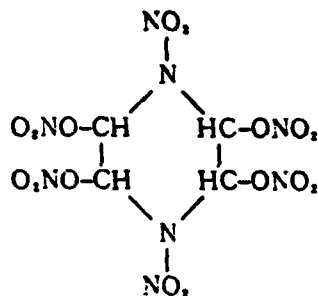
Tseng-hwei Wang and Deyu Tian designed and synthesized the 2,3,5,6-Tetranitro-1,4-Dinitropiperazine (TN) in the early seventies. When compared with commonly used oxidizer Ammonium Perchlorite (Ap), TN has the advantages such as it does not contain chloride in the element; high energy. All of these have never been reported before. This paper briefly describes the physical and chemical properties of TN. Based on White's Minimization of Gibbs Free Energy theory, FORTRAN language was used to program a series of computer routines. Calculations were conducted on DPS-6 system. Systematic computations were performed on HTPB (Hydroxyl Terminated Poly Butadiene

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) -TN, HTPB-Al-TN and HTPB-Al-Ap-TN systems of composite solid propellants. Parameters such as theoretical impulse (I_{sp}), characteristic speed (c^*), combustion chamber temperature (T_c), and combustion products were found. Definite routines have also been derived. When the weight ratio of TN to HTPB is 9 to 1 in the HTPB-TN propellants, the theoretic impulse reaches its maximum value of 269.33 seconds. Energetic characteristic value increases with the increase of TN content in the HTPB-TN-Ap-A propellant. Should TN replace all Ap, I_{sp} can be increased by ten seconds. It is obvious that TN definitely is a high energetic organic oxidizer. It deserves further study.

2. CONCISE PHYSICAL AND CHEMICAL PROPERTIES OF 2,3,5,6-TETRANITRATO-1,4-DINITROPIPERAZINE (TN)

TN is a new type of high energy organic oxidizer. Synthesized TN is in the form of white short cylindrical crystal. Melting point is 138-140 degrees celcius. Through element analysis, infrared spectrum and nuclear-magnetic resonance its chemical formula is determined to be $C_4H_4N_8O_{16}$ with the configuration:



Heat of combustion is -4647.17 kJ/kg, Heat of formation is -220.62 kJ/mole (-525.26 kJ/kg). Specific weight d_{20}^{20} is 1.812. Theoretic oxygen content is 60.95%. Oxygen content per unit volume is 1.105 g/cm³, close to liquid oxygen (for liquid oxygen, the oxygen content per unit volume is only 1.14 g/cm³).

3. ENERGETIC CHARACTERISTIC OF TN IN SOLID PROPELLANT

Based on White's minimization of Gibbs Free Energy theory⁽¹⁾, FORTRAN language is used to program the computer sequence to calculate the propellant's energy characteristics. Parameters such as theoretic impulse I_{sp} , Combustion Chamber Temperature T_c , and average molecular weight of gaseous products M_{sp} are computed. Results from calculation were close to documented values⁽²⁾. Theoretic impulse fluctuates within the range of one second, see table 1. This shows that the calculation method and sequence are accurate and reliable.

The discussion in this paper is restricted to a simple propellant system composed of oxidizer, adhesives and metallic additives. It is assumed that other minor components do not affect the energy greatly. Properties of compounds used can be seen in table 2.

HTPB was chosen to be the adhesive. Computation of the energetic characteristic values were conducted for the following three systems:

1. The effects of TN and Al content changes on energetic characteristics in the TN-HTPB-Al system. HTPB content is fixed (11%), the energy of the propellant increases with the increase of aluminum content (TN content is reduced accordingly). Aluminum content increased by 20%, theoretical impulse increased seven seconds, combustion chamber temperature increased by 680K while characteristic

velocity is increased by 48 meter/sec (See Table 3 and Figure 1). Calculation results show that with the increase of combustion chamber temperature T_c , theoretic impulse I_c , and characteristic velocity C_c , both increase. However, with the increase of the average molecular weight of combustion products, the theoretic impulse and characteristic velocity both decrease. The insulation index does not have great influence. All this agrees with the following equation.

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$$I_c \propto \sqrt{T_c / M_c}$$

where M_c is the average molecular weight of gaseous products.

2. The influence of TN and Ap content changes in the TN-Ap-Al-HTPB system /87
on the energetic characteristics

When the HTPB and Al contents were fixed (HTPB was 11%, Al was 16%), the energy of the propellant increased with the increase of TN content (Ap content decreased correspondingly). On the average when the content of TN increases by 1%, the specific impulse increases by 0.15 second. The combustion chamber temperature increases by 5.6K, while the characteristic speed increases by 0.93 meter/second. If the Ap in the system composition were totally substituted by TN, the I_{sp} would exceed those propellants without TN by approximately ten seconds. The results from calculations can be seen in table 4 and figure 2.

It was well known that the energy content of a propellant is proportional /88 to the summation of its components' formation heats. Because:

$$\Delta H_{\text{combustion}} = \Delta H_{\text{product}} - \Delta H_{\text{propellant}}$$

where $\Delta H_{\text{combustion}}$, $\Delta H_{\text{product}}$ and $\Delta H_{\text{propellant}}$ are the heat of combustion of the propellant, formation heat of the products and the formation heat of the propellant components respectively. The formation heat of TN is greater than that of Ap, therefore the increase of TN content of TN results in the enhancement of energy content. From the chemical configuration point of view, TN has more high-energy radical groups $-\text{NNO}_2$, $-\text{ONO}_2$. TN does not contain the element Cl, therefore the energy of TN is greater than that of Ap. Calculations also verified this point.

3. The influence of TN and HTPB content changes in the TN-HTPB smokeless propellant system on the energetic characteristics

With the increase of TN content (HTPB content decreases correspondingly), the theoretic specific impulse (I_{sp}), characteristic speed (C^*) both increase promptly. TN increases from 80% to 90%. This results in the increase of I_{sp} from 247.96 second to 269.33 second. I_{sp} increased by 21.4 seconds. However, when the content of TN kept on increasing, the energetic characteristic values such as I_{sp} started to decrease gradually.

When the TN content was 90%, theoretic specific impulse I_{sp} had the maximum value. See table 5 and figure 3 for this result. At the maximum point, the oxygen equilibrium for the propellant was -11.4%. /88

Viewed from the calculation results, with the increase of TN content, the CO_2 content in the product increases promptly. H_2O and CO contents decrease at the same time. The heat released through combustion increased, the combustion temperature increased also, therefore the specific impulse was enhanced. However, when the TN content was too high, the degree of oxidization of the products increased, i.e. the contents of CO_2 and O_2 both increased; when TN content exceeds 90%, the content of O_2 increased significantly which resulted in the decrease of combustion temperature. Average molecular weight of the product increased because of the complicated molecules such as CO_2 and O_2 increased greatly in the products. Specific impulse, therefore, decreased promptly. See table 6 and figure 4 for this phenomenon.

The composite smokeless propellant composed by HTPB and TN does not contain elements Al and Cl. The smoke generated by combustion is greatly reduced. This meets the requirements for practical applications on satellites, cosmic aeroships and tactical rocket engines. When compared with propellants containing Al or Cl, it has the advantages such as low combustion temperature, small combustion erosion on the engine nozzle, low average molecular weight of combustion gases, no two-phase flow losses and high energy efficiency. /89

Because the effective oxygen content of TN is high and also its energy content is high, therefore we have tried to use TN to replace the oxidizer Ammonium nitrate in non-smoke propellant. Calculation research has been performed on that system. The results showed that with the addition of TN, the specific impulse of that non-smoke system was greatly enhanced. The highest specific impulse was close to those propellants containing Aluminum and Ap. From the above calculations and discussions, theoretically speaking, TN is an energy-containing organic oxidizer.

4. CONCLUSION

Nitrogen content of 2,3,5,6-Tetranitrato-1,4-Dinitropiperazine is 26.67%. Oxygen content (61%) is high. Density is relatively large. Oxygen content per unit volume (1.105 g/cm^3) is close to that of liquid oxygen. Theoretic specific impulse for single-component propellant is 233.21 seconds. When mixed with HTPB to form smokeless propellant, the specific impulse increases with the increase of TN content. When TN content reaches 90%, the theoretic specific impulse has the maximum value of 269.33 seconds. Characteristic speed C^* reaches 1610.5 meter/sec. The composite solid propellant system composed of TN, HTPB and Al has theoretic specific impulse of 276 seconds with characteristic speed 1662 meter/second.

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For the HTPB-Al-TN-Ap propellant system, when HTPB, Al contents are fixed, specific impulse increases with the increase of TN content (Ap content decreases correspondingly). The value changes between 265 and 275 seconds.

We arrive at the conclusion from above discussion: when energy-containing oxidizer and other energy-containing module of the propellant are employed, the overall energy content is enhanced.

This paper starts from the discussion of energy. Theoretic contributions of TN as an oxidizer to propellant energy is examined. Calculation results show that since TN's oxygen content is higher than that of Ap, also TN is an energy-containing oxidizer, therefore, using TN as a replacement of Ap to be the oxidizer enhances the energy content of the propellant. The fact that TN does not contain Cl element makes TN a candidate to be used in the smokeless propellant system.

The authors wish to thank Associate professor Jiang Yu's assistance and instruction.

REFERENCES

- [1] W. B. White et al J. Chem. Phys, Vol. 28, p. 751, 1958.
- [2] Barrel Siegeland and Leory Schieler, Energetic of Propellant Chemistry, 1964.

表1 用该程序计算的能量示性数与文献值⁽¹⁾的比较

(2) 序号	(3) 配 方 (%)			(4) 计算方法	(5) 能 量 示 性 数		
	Ap	PV	Al		$I_{sp}(S)$	$T_e(K)$	\bar{M}_e
1	95	5	—	(6) 本 法	219.58	2541.51	27.45
				(7) 文献法	219	2529	27.47
2	90	10	—	(6) 本 法	252.16	3014.8	28.10
				(7) 文献法	252	3013	26.13
3	80	20	—	(6) 本 法	221.06	2054.08	20.43
				(7) 文献法	221	2036	20.43
4	81	9	10	(6) 本 法	280.99	3432.9	28.52
				(7) 文献法	261	3433	28.54
5	78.5	8.5	15	(6) 本 法	263.82	3626.8	29.81
				(7) 文献法	264	3621	29.83

注: PV为聚乙烯 (6)

Table 1 Comparison of calculated energetic characteristic values by computer program and documented value^(L)

- (2) Serial number (6) Our method
 (3) Composition (7) Documented method
 (4) Calculation method (8) Note: PV stands for polyethylene
 (5) Energetic characteristic values

表2 所用化合物的简要性能⁽¹⁾

(2) 化 合 物 名 称	(3) 代 号	(4) 化 学 式	(5) 密 度 (g/cm ³)	(6) 生 成 热	
				kJ/kg	kcal/kg
(7) 2,3,5,6-四硝基肼-1,4-二硝基吡嗪	TN	C ₄ H ₄ N ₈ O ₁₄	1.812	-525.26	-125.54
(8) 过 氯 酸 铵	Ap	NH ₄ ClO ₄	1.95	-2471.95	-590.81
(9) 铝 粉	Al	Al	2.7	0	0
(10) 端羟基聚丁二烯	HTPB (R-45M)	C ₄ H _{6.03} O _{0.03}	0.93	-19.04	-4.55
(11) 聚 乙 烯	PV	(CH ₂) _n	0.92	-1917.95	-458.4

Table 2 Basic properties of the compounds employed

- (2) Name of the compound (7) Polyethylene
 (3) 2,3,5,6-Tetranitrato-1,4-Dinitropiperazine (8) Code name
 (4) Amonium Perchlorite (9) Chemical formula
 (5) Aluminum powder (10) Density
 (6) Hydroxyl-Terminated Poly Butadiene (11) Heat of formation

表3 TN 与 Al 不同含量对推进剂能量示性数的影响

(2) 序号	(3) 配 方 (%)			(4) 能 量 示 性 数				
	TN	Al	HTPB	$I_{sp}(S)$	$T_c(K)$	\bar{M}_{gc}	$C^*(m/s)$	k
1	89	0	11	269.04	3475.4	26.96	1614.2	1.1640
2	84	5	11	271.34	3646.1	25.20	1629.7	1.1597
3	79	10	11	273.42	3821.6	23.30	1644.2	1.1559
4	74	15	11	275.22	3999.6	21.30	1657.5	1.1526
5	72	17	11	275.83	4066.2	20.49	1661.8	1.1511
6	69	20	11	276.01	4155.6	19.55	1661.9	1.1450

(5) 注: \bar{M}_{gc} 为产物中气体的平均分子量

Table 3 Effects of different contents of TN and Al on the energetic characteristic values of the propellants

- (2) Serial number
 (3) Composition
 (4) Energetic characteristic value
 (5) Note: \bar{M}_{gc} stands for the average molecular weight of the gaseous product

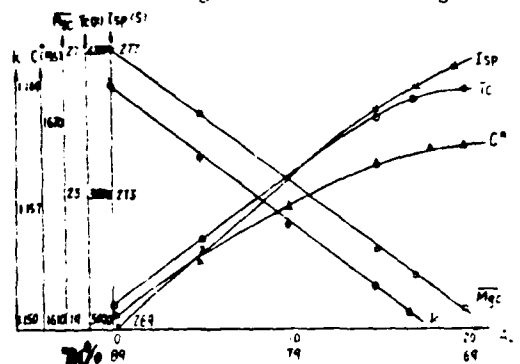


图1 TN 和 Al 含量变化对能量特性的影响
 Figure 1 Influence of different contents of TN and Al on the energetic characteristic values

表4 TN 与 Ap 不同含量对推进剂能量示性数的影响⁽¹⁾

(1) 序号	(2) 配 方 (%)				(3) 能 量 示 性 数			
	TN	Ap	Al	HTPB	$I_{sp}(S)$	$T_c(K)$	O.B(%)	$C^*(m/s)$
1	70	3	16	11	275.05	4012.3	-32.34	1656.6
2	65	8	16	11	274.25	3978.1	-31.79	1651.4
3	60	13	16	11	273.47	3945.6	-31.23	1646.3
4	50	23	16	11	271.95	3884.5	-30.11	1636.6
5	40	33	16	11	270.50	3827.2	-28.99	1627.8
6	30	43	16	11	269.04	3773.4	-27.87	1617.8
7	20	53	16	11	267.58	3722.1	-26.75	1608.8
8	10	63	16	11	266.12	3672.6	-25.63	1600.0
9	5	68	16	11	265.38	3648.5	-25.07	1595.6
10	0	73	16	11	264.60	3624.6	-24.51	1591.3

Table 4 Influence of different contents of TN and Ap on the energetic characteristic values of the propellants

- (2) Serial number
- (3) Composition
- (4) Energetic characteristic value

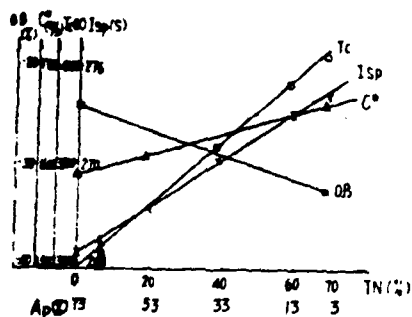


图2 TN 与 Ap 含量变化对能量特性的影响

Figure 2 Influence of different contents of TN and Ap on the energetic characteristics

表5 TN-HTPB 系列无烟推进剂的能量示性数⁽¹⁾

(2) 序号	(3) 配 方 (%)		(4) 能 量 示 性 数			
	TN	HTPB	$I_{sp}(s)$	$T_c(K)$	$C^*(m/s)$	O.B(%)
1	100	0	233.21	3086.3	1405.0	22.85
2	95	5	258.74	3401.5	1543.5	5.7
3	92	8	267.30	3477.1	1595.2	-4.5
4	91	9	268.79	3486.0	1604.8	-7.9
5	90	10	269.33	3485.9	1610.5	-11.4
6	89	11	269.04	3475.4	1614.2	-14.8
7	88	12	268.10	3453.0	1616.1	-18.2
8	86	14	264.75	3356.5	1613.5	-25.1
9	84	16	260.05	3217.8	1600.9	-31.9
10	82	18	254.39	3024.6	1578.6	-38.8
11	80	20	247.96	2805.9	1548.1	-45.6

Table 5 Energetic characteristic values of TN-HTPB smokeless propellant system

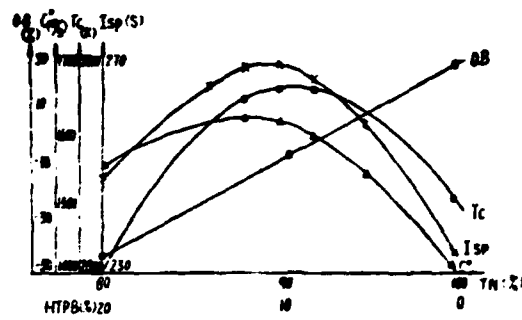
- (2) Serial number
(3) Composition
(4) Energetic characteristic value

表6 TN-HTPB系列无烟推进剂的比冲及燃烧产物

(2) 序号	(3) 配 方 (%)		(4) 比 冲	(5) 燃 烧 产 物 (mole/kg)				
	TN	HTPB		CO	CO ₂	H ₂ O	O ₂	N ₂
1	100	0	233.21	1.0001	8.5198	4.2584	6.8104	8.9924
2	95	5	258.74	4.1382	8.5448	6.0690	3.0199	8.5707
3	92	8	267.30	6.7658	7.8144	7.0856	1.4945	8.4056
4	91	9	268.79	7.7479	7.4646	7.3990	1.1078	8.3580
5	90	10	269.33	8.7820	7.0630	7.6990	0.7837	8.3122
6	89	11	269.05	9.8663	6.6110	7.9485	0.5228	8.2666
7	88	12	268.10	10.9972	6.1125	8.1601	0.3241	8.2191
8	86	14	264.75	13.3606	5.0136	8.3628	0.0939	8.1095

Table 6 Specific impulse and combustion product of TN-HTPB smokeless propellant system

- (2) Serial number
(3) Composition
(4) Specific impulse
(5) Combustion product



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图3 HTPB与TN含量的变化对能量特性的影响
Figure 3 Influence of different contents of HTPB and TN on energetic characteristics

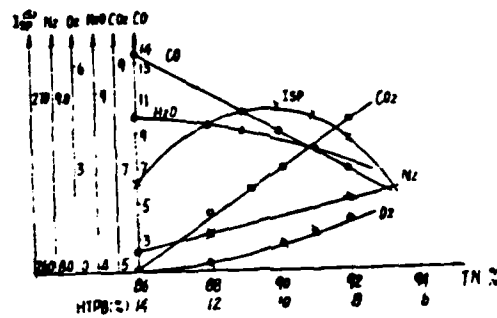


图4 HTPB与TN含量变化对比冲及燃烧产物的影响
Figure 4 Influence of different contents of HTPB and TN on specific impulse and combustion product

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